Raman scattering of light by electrons in superconductors: contribution of surface region and Coulomb-interaction effects

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Raman scattering (RS) of light by electron excitations of a superconductor (SC) is considered. A longitudinal electric field is induced in a metal near its boundary (at a depth on the order of the screening length) by an (incident or scattered) field component normal to the SC boundary. It is shown that in a superconductor with an isotropic Fermi surface the heretofore considered contribution made to the RS cross section by longitudinal fields exceeds by several orders the contribution of the transverse (penetrating to the skin-layer depth) field components.

1. INTRODUCTION

In connection with the discovery of high-temperature superconductors (SC), interest has revived in the interaction between an SC and an electromagnetic field, particularly in Raman scattering (RS) of light with excitation of electron–hole pairs. The theory of RS in the BCS model was developed in Refs. 1–4, where the interaction of metal electrons with transverse components of the incident and scattered electromagnetic fields was studied. The amplitudes of these components attenuate in the metal to a skin-layer depth δ ($\sim 10^2$–10^3 Å), and the main contribution to the RS cross section is determined by the diagram of Fig. 1, i.e., by the imaginary part of the density–density correlator. In the case of an isotropic SC, the Coulomb screening suppresses the long-wave fluctuations of the electron density, so that the cross section for RS in an isotropic single-band SC, due to the transverse field components, is extremely small. For an anisotropic electron spectrum, the effects of the Coulomb screening are not so significant,[2,4] and the RS cross section determined by the diagram of Fig. 1 makes it possible to interpret the experimental data for certain SC.[5]

Note, however, that in RS experiments one encounters usually both tangential and normal components of electromagnetic fields. Even when the incident light is s-polarized, scattered light gathered in a large scattering angle (weak signal) has a p-polarized component. In turn, the presence of p-polarized light produces a longitudinal electric field in the metal near its surface. The amplitude of this field attenuates rapidly (over a distance on the order of the Thomas–Fermi screening radius k_{TF}^{-1}). The role of processes with large momentum transfers becomes substantial in this case and, as will be shown below, the contribution made to the RS cross section by longitudinal fields exceeds in an isotropic SC the contribution of the transverse field components by several orders.

2. CONTRIBUTION OF LONGITUDINAL FIELD COMPONENTS TO RAMAN SCATTERING OF LIGHT

We separate in the considered inhomogeneous (semi-bounded) medium the longitudinal and transverse components of the incident or scattered field in the following manner:

$$\mathbf{E}\equiv\mathbf{E}_l+\mathbf{E}_t=-\nabla\varphi-\frac{1}{c}\frac{\partial\mathbf{A}}{\partial t}, \quad (1)$$

$$\operatorname{div}\mathbf{A}=0. \quad (2)$$

The gauge (2) is suitable for the present problem, where nonlocality (spatial dispersion) of the electromagnetic response of the metal electrons plays an important role. For an abrupt boundary ($z=0$) of media without spatial dispersion, the z component of the electromagnetic field in the usual Fresnel problem is of course singular, having a discontinuity at $z=0$. This, however, does not lead to singular consequences, for in the "interior" of the medium, i.e., in our case already at $z\neq0$, the field varies smoothly, particularly in a metal, where it attenuates within the skin-layer δ. If nonlocality is present, the singularity is smoothed out, and then the longitudinal field $\varphi(\mathbf{r})$ due to the appearance of a surface charge near the interface differs from zero inside the metal down to depths of order k_{TF}^{-1} (charges are produced near the surface if the $\mathbf{E}(\mathbf{r})$ component normal to the surface differs from zero). At the same time, the skin-layer depth remains the scale of variation of the transverse field $\mathbf{A}(\mathbf{r})$ in the metal. By assuming the electrons to be specularly reflected from the metal boundary we can, following the procedure of Ref. 6, solve completely the linear problem of determining the field $\mathbf{E}(\mathbf{r})$ inside the metal with allowance for spatial dispersion.

Let us estimate the contribution of the longitudinal field component ($-\nabla\varphi$) to the RS cross section. The Hamiltonian of the interaction of this field with the electron is

$$H_{int}=e\int\rho(\mathbf{r})\varphi(\mathbf{r})d\mathbf{r},$$

where $\rho(\mathbf{r})$ is the electron-density operator. Neglect of the transverse field component ($\sim\mathbf{A}$) corresponds to neglect of the retardation, the cause of which, as will be shown below, is that the main contribution to the RS cross section is made by processes with large momentum transfer ($\sim k_{TF}$; this is characteristic scale of variation of the potentials $\varphi_{i,s}(\mathbf{r})$ of the incident and scattered fields). In contrast to the usually considered interaction with a transverse field, when the

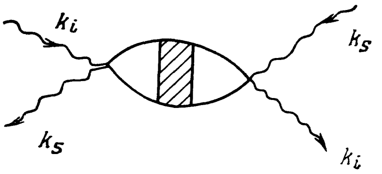

FIG. 1.

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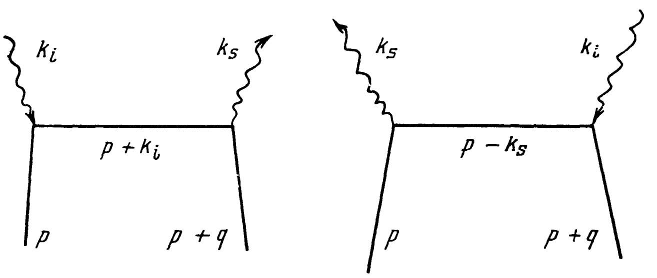

FIG. 2.

main contribution to the scattering amplitude M is determined by first-order perturbation theory in $H'_{\rm int}$:

$$H'_{int}=\frac{e^2}{2mc^2}\int \mathbf{A}^2(\mathbf{r})\rho(\mathbf{r})\,d\mathbf{r},$$

a nonzero contribution appears in this case only in second order in $H_{\rm int}$ (Fig. 2). Since $k_{\rm TF}\gg\xi^{-1}$ (ξ is the SC coherence length), we neglect the corrections to M due to the pairing BCS interaction of the quasiparticles in the final state[4,7]; these corrections are significant only in a narrow frequency region $\omega_i-\omega_s\equiv\omega=2\Delta$ near the threshold. Assuming the high-density assumption $k_{\rm TF}\ll p_F$ to be valid (p_F is the Fermi momentum), we neglect also the correlation corrections to M by the Coulomb interaction. Nor will account be taken of Coulomb screening effects, which do not lead to a substantial change of the estimates obtained below as a result of the relatively large momentum transfer.

The RS differential scattering cross section (the flux ratio of the scattered field to the field incident at the angle θ_i) is of the form

$$d\sigma(\omega)=\int\frac{dq}{2\pi}\int\frac{d^3p}{(2\pi)^3}|M(\mathbf{p},\mathbf{q})|^2\delta[E_\mathbf{p}+E_{\mathbf{p}+\mathbf{q}}-\omega]\times\frac{\omega_s^2\,d\omega\,d\Omega}{(2\pi)^3\cos\theta_i},\tag{3}$$

where

$$E_\mathbf{p}=(\varepsilon_\mathbf{p}^2+\Delta^2)^{1/2},\quad \varepsilon_\mathbf{p}=\mathbf{p}^2/2m-\varepsilon_F,$$

$$M(\mathbf{p},\mathbf{q})=e^2\int\frac{dk_i\,dk_s}{2\pi}\varphi_s^*(k_s)F(k_i,k_s,\mathbf{p})\varphi_i(k_i)\delta(k_i-k_s-q).\tag{4}$$

Equations (3) and (4) were derived by the usual[1,6] procedure of specular continuation of the fields $\varphi(\mathbf{r})$ into all of space (assuming specular electron reflection from the boundary). This is justified here by the inequality $k_{\rm TF}\ll p_F$ (i.e., the electron wavelength is small compared with the scale of variation of the potentials $\varphi_{i,s}(\mathbf{r})$. Since the potentials $\varphi_{i,s}(\mathbf{r})$ vary smoothly along the interface and very rapidly along the normal to it (the z axis), we need naturally allow for the dependences of $\varphi_{i,s}$ on only the coordinate z—this is in fact reflected in the one-dimensional character of the integration with respect to q, k_i, and k_s in (3) and (4).

The form factor $F(k_i,k_s,\mathbf{p})$ is

$$F(k_i,k_s,\mathbf{p})=\left[\frac{A_i}{\omega_i-E_\mathbf{p}-E_{\mathbf{p}+\mathbf{k}_i}}-\frac{B_i}{\omega_i-E_\mathbf{p}+E_{\mathbf{p}+\mathbf{k}_i}}\right]+\left[\begin{array}{c} i\to s\\ \omega_i\to-\omega_s\\ \mathbf{k}_i\to-\mathbf{k}_s\end{array}\right],\tag{5}$$

where

$$A_i=R^-(\mathbf{p},\mathbf{p}+\mathbf{k}_i)R^+(\mathbf{p}+\mathbf{k}_i,\mathbf{p}+\mathbf{q}),\quad A_s=A_i(\mathbf{k}_i\to-\mathbf{k}_s),$$
$$B_i=R^+(\mathbf{p},\mathbf{p}+\mathbf{k}_i)R^-(\mathbf{p}+\mathbf{k}_i,\mathbf{p}+\mathbf{q}),\quad B_s=B_i(\mathbf{k}_i\to-\mathbf{k}_s).$$

The coherence factors $R^\pm$ are equal to

$$R^+(\mathbf{k},\mathbf{k}')=\cos\frac{\psi(\mathbf{k})+\psi(\mathbf{k}')}{2},$$

$$R^-(\mathbf{k},\mathbf{k}')=\sin\frac{\psi(\mathbf{k})+\psi(\mathbf{k}')}{2},$$

$$\cos\psi(\mathbf{k})=\varepsilon_k/E_\mathbf{k},\quad \sin\psi(\mathbf{k})=\Delta/E_\mathbf{k},\quad 0<\psi<\pi.$$

In the case of interest to us, when $2\Delta\lesssim\omega\ll\omega_{i,s}$, the energy conservation law expressed by the delta-function in (3) restricts the energies $E_\mathbf{p}$ and $E_{\mathbf{p}+\mathbf{q}}$ to a region near the Fermi surface: $E_\mathbf{p}$, $E_{\mathbf{p}+\mathbf{q}}\lesssim\omega\ll\varepsilon_F$. Owing to the large value of the transferred-momentum component, q_z, this leads in turn to the approximate equality $p_z\approx\pm q_z/2$. At the same time, since the momenta $k_{i,s}$ are large, the intermediate-state energies are large:

$$E_{\mathbf{p}+\mathbf{k}_i},\ E_{\mathbf{p}-\mathbf{k}_s}\sim\varepsilon_F,$$

meaning that

$$E_{\mathbf{p}+\mathbf{k}_i}\approx|\varepsilon_{\mathbf{p}+\mathbf{k}_i}|,\quad E_{\mathbf{p}-\mathbf{k}_s}\approx|\varepsilon_{\mathbf{p}-\mathbf{k}_s}|.$$

With allowance for the relation indicated for p_z, we have

$$|\varepsilon_{p+k_i}|\approx|\varepsilon_{p-k_s}|\approx|k_{iz}k_{sz}|/2m.$$

These relations lead to the explicit expression (4) for $M(\mathbf{p},\mathbf{q})$. This expression is particularly simple in the static limit (i.e., when the frequencies $\omega_{i,s}$ in (5) are neglected compared with $E_{\mathbf{p}+\mathbf{k}_i}$ and $E_{\mathbf{p}-\mathbf{k}_s}\sim\varepsilon_F$. Confining ourselves for simplicity to this approximation, we get

$$\frac{d\sigma}{d\omega}=\frac{16\pi m^2e^4}{\cos\theta_i}\int\frac{dq}{2\pi}\int\frac{d^3p}{(2\pi)^3}\left(1-\frac{\varepsilon_\mathbf{p}\varepsilon_{\mathbf{p}+\mathbf{q}}-\Delta^2}{E_\mathbf{p}E_{\mathbf{p}+\mathbf{q}}}\right)\times\delta(E_\mathbf{p}+E_{\mathbf{p}+\mathbf{q}}-\omega)\times\left|\int\frac{dk_i\,dk_s}{2\pi}\frac{\varphi_s^*(k_s)}{k_s}\frac{\varphi_i(k_i)}{k_i}\delta(k_i-k_s-q)\right|^2\frac{\omega_s^2\,d\Omega}{(2\pi)^3}.\tag{6}$$

The expression for the potentials of the longitudinal electric field takes in the static limit the form

$$\varphi_{i,s}(z)=\varphi_{i,s}(0)\exp[-k_{TF}|z|]$$

(with account taken of the specular continuation), where the potentials $\varphi_{i,s}(0)$ on the boundary are expressed in terms of the photon amplitudes of the incident and scattered p-polarized plane waves outside the metal:

$$\varphi_{i,s}(0)=\frac{2E_{i,s}}{k_{TF}}\sin\theta_{i,s},$$

where θ_i (θ_s) is the incidence (scattering) angle.

The integral with respect to q in (6) takes for $q\gtrsim\xi^{-1}\sim\max\{\Delta/v_F,\omega/v_F\}$ the form

$$\int_{\xi^{-1}}^{2p_F}\frac{1}{[q^2+(2k_{TF})^2]^2}\frac{dq}{q},$$

i.e., it is determined by values of q that are large compared with ξ^{-1}. The contribution made to (6) by the region of small q is significant only near the threshold $\omega - 2\Delta \ll \Delta$.

The final expression for the cross section is

$$d\sigma = \frac{2^9}{\pi^2} \frac{\sin\theta_i \sin^2\theta_s}{\cos\theta_i} \frac{e^4 m^4 \omega_i \omega_s^3 \omega^2}{\hbar^6 c^4 (k_{TF})^{10}} \times \ln(k_{TF}\xi) E\left(\left(1-\frac{4\Delta^2}{\omega^2}\right)^{1/2}\right) \frac{d\omega}{\omega} \frac{d\Omega}{4\pi}. \qquad (7)$$

We have restored here the constants $\hbar$ and c; the meaning of ξ in this case is $\xi = \max\{\omega/v_F, \Delta/v_F\}$ and $E(x)$ is a complete elliptic integral.

3. DISCUSSION OF RESULTS

We note first of all that in isotropic SC the value of d [Eq. (7)] exceeds by several orders the transverse-field scattering cross section $d\sigma_\perp$, the expression for which at $v_F/\Delta \sim \delta$, where δ is the skin-layer depth, is

$$d\sigma_\perp \sim \frac{e^4\omega^2}{\varepsilon^2\delta^2 k_{TF}^2 \hbar^2 c^4} \ln(k_{TF}\delta) E\left(\left(1-\frac{4\Delta^2}{\omega^2}\right)^{1/2}\right) \frac{d\omega}{\omega} \frac{d\Omega}{4\pi}, \qquad (8)$$

where ε is the dielectric constant of the metal at the incident-light frequency ($|\varepsilon| \sim \omega_p^2/\omega_i^2$, ω_p is the plasma frequency). Expression (8) differs from the one obtained in Ref. 1 with allowance for screening in the density–density correlation function. Putting $k_{TF} \sim p_F$, we easily obtain the relation

$$d\sigma_\perp / d\sigma \sim (k_{TF}\delta)^{-2} \sim 10^{-5}.$$

An estimate of $d\sigma$ [Eq. (7)] for $\hbar\omega_{i,s} = 2.5$ eV and $\hbar\omega \sim 2\Delta = 4\cdot 10^{-3}$ eV yields

$$d\sigma \sim 10^{-16} (k_{TF}\cdot\text{Å})^{-10} \frac{d\omega}{\Delta} \frac{d\Omega}{4\pi}. \qquad (9)$$

Assuming, for example, $k_{TF} = 0.8$ Å^{-1}, we obtain for $d\sigma$ an estimate of the same order as in experiment.[5] This estimate is made less reliable, however, by the strong dependence of $d\sigma$ on k_{TF} under conditions when the random-phase approximation is valid in real metals. Thus, for example, for $k_{TF} = 1$ Å^{-1} we find $d\sigma$ to be smaller by an order of magnitude, indicating that such comparisons are somewhat arbitrary. There is no doubt, however, that the considered scattering channel predominates in isotropic semiconductors.

The investigated scattering mechanism can thus be regarded in principle as an alternative (not related to anisotropy effect) possibility of explaining the relative large RS in SC. It is not excluded that this mechanism can become significant also in the interpretation of RS results for high-temperature SC which, however, are not good "dense" metals. It is also noteworthy that the cross section is determined by the contribution of large momentum transfers and, hence, by a narrow surface region (on the order of several Å).

Experimental proof of the proposed mechanism would be a strong dependence of the RS cross section on the polarizations of the incident and scattered radiation.

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